Expert Report of Joel D Blum

Confederated Tribes of the Colville Reservation v. Teck Cominco Metals Ltd.

Prepared for Short Cressman & Burgess PLLC, on behalf of the Confederated Tribes of the Colville Reservation and the State of Washington.

Report Prepared August 1, 2014

Prepared by:
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(b)(6)

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Joel D Blum, August 1, 2014

1. INTRODUCTION

1.1 Retention and Assignment

On behalf of the Confederated Tribes of the Colville Reservation (Tribes) and the State of Washington I have been retained by Short, Cressman & Burgess PLLC, counsel for the Tribes, to prepare an Expert Report summarizing my analysis of whether metals from the Teck-Cominco Trail Smelter were emitted from the Trail Smelter stacks, transported in the atmosphere and deposited onto lakes, forests and other lands of the Upper Columbia River Valley and adjacent uplands south of the Canadian Border (the UCR site). I am being compensated for my work on this case at the rate of $200/hour for office and field work and $400/hour for deposition and trial testimony.

The opinions presented in this report are based on my review of pertinent reports, data, publications and other documents provided to me by Short, Cressman & Burgess PLLC and obtained by me from the publically accessible literature. Mercury concentrations and stable isotopic compositions for sediments from Lakes Bonaparte, Ellen and Cedar, that are presented herein, were measured by Dr. James Gleason at the University of Michigan. Dr. Gleason has over 2 years of experience with high precision concentration and isotope ratio analyses. The conclusions presented in this report represent my current opinions concerning the scientific issues that are addressed.

1.2 Qualifications

have been a tenured Full Professor at the University of Michigan since 1999 and have appointments in the Departments of Earth and Environmental Sciences and Ecology and Evolutionary Biology at the University of Michigan (Ann Arbor, MI). I am the holder of the University of Michigan's endowed John MacArthur Professorship and I am a past Department Chairman. I received my PhD in Geochemistry in 1990 from the California Institute of Technology (Pasadena, CA) and was Professor of Earth Sciences at Dartmouth College (Hanover, NH) from 1990 to 1999 before being recruited to the University of Michigan.

My expertise is in the study of trace metals in the environment. During the past 15 years I have focused much of my effort on research related to the geochemical and biogeochemical behavior of mercury. I have published approximately 185 research articles and my work has been cited over 8300 times. I have received over 40 research grants amounting to over $7M in research funding. I have served on numerous advisory panels for US funding agencies including the NSF, NIH, DOE and EPA. I am the past Editor in Chief of the journal Chemical Geology, and I am presently Editor in Chief of the journal Elementa. I am a Fellow in the American Association for the Advancement of Science, the Geochemical Society, the American Geophysical Union and the Geological Society of America. In 2013 I was awarded the Patterson Award by the Geochemical Society "for outstanding contributions to environmental geochemistry." I have provided expert opinions in two previous cases "Confederated Tribes of the Colville Reservation v. Teck Cominco Metals Ltd" and "Clipper Yacht Harbor v. City of Sausalito." A full list of my publications over the past ten years are included in Appendix A.

2. STATEMENT OF OPINIONS

2.1 **OPINION 1**

Mercury concentration profiles in sediment cores from Lake Bonaparte, Lake Ellen and Cedar Lake in northeastern Washington State provide a spatial and temporal record of mercury deposition from the atmosphere in the Upper Columbia River Valley an show that mercury deposition was greatest in close proximity to the Trail smelter.

2.2 **OPINION 2**

Mercury stable isotope ratios in sediments from Lake Bonaparte, Lake Ellen and Cedar Lake provide isotopic fingerprints of changing proportions of mercury between ore deposits (or smelters that process the ores) and regional/global background atmospheric sources. This demonstrates that mercury in Cedar Lake sediments is derived primarily from ore deposit (or smelter) sources, and that about half of the mercury in Lake Ellen sediments is derived from ore deposit (or smelter) sources.

2.3 **OPINION 3**

Historical data concerning Trail smelter SO_2 emissions and meteorological analysis of the transport of its emissions corroborates and provides independent evidence for my opinions identifying atmospherically deposited metals from the Trail smelter in the Upper Columbia River Valley site.

3. BASES OF OPINIONS

3.1 **OPINION 1**

Mercury concentration profiles in sediment cores from Lake Bonaparte, Lake Ellen and Cedar Lake in northeastern Washington State provide a spatial and temporal record of mercury deposition from the atmosphere in the Upper Columbia River Valley an show that mercury deposition was greatest in close proximity to the Trail smelter.

LOCATION AND PROPERTIES OF STUDY LAKES

Three lakes were chosen for sediment coring to provide spatial and temporal information on atmospheric deposition of metals in the Upper Columbia River Valley and adjacent uplands south of the Canadian Border (the UCR site). Detailed descriptions of coring methods and sediment dating using ^{210}Pb and ^{137}Cs profiles are given in Vlassopoulos (2014). Briefly, lakes were chosen with remote locations, small surface area (50-150 acres), and small catchment areas (≤ 1 times the lake surface area) to minimize inputs from watershed run-off compared to atmospheric inputs of metals to the lake surfaces. Lake Bonaparte was selected as a reference lake for lake sediment sampling because it is separated from the Columbia River Valley by several mountain ranges (including the Kettle Mountains); it is 105 km southwest from Trail and 7 km west of the upper Columbia River Valley. Cedar Lake and Lake Ellen are situated 2 km and 8 km from Trail BC in the UCR. Cedar Lake is the closest lake to Trail, BC that met our lake selection criteria and is in the zone of visible smelter plume injury to forest vegetation that was mapped in the years 1929-1931 (USDA 1936). Lake Ellen is about 1 km south of the mapped 1929-1931 forest injury area and together these lakes provide spatial gradient in the predicted

deposition of atmospheric pollutants from the Trail smelter (Haney 2011). Gaseous and fine particulate forms of sulfur and mercury would be expected to follow similar atmospheric transport patterns and to have shown enhanced deposition in areas of visible smelter plume injury to forest vegetation (Haney 2011).

ESTABLISHMENT OF BACKGROUND LAKE SEDIMENT MERCURY CONCENTRATIONS

Sediments were dated using ^{210}Pb and ^{137}Cs profiles and sediment accumulation rates were estimated for each lake. Methods used for estimating the year of deposition of each sediment core slice (and associated uncertainties) are discussed in Vlassopoulos (2014). Calendar years are assigned to each core slice but sediment mixing as well as Pb and Cs mobility result in uncertainties of u to a decade in year of deposition (Vlassopoulos 2014). The average mercury concentration of sediments deposited before the era of industrial activity (pre-anthropogenic; from 1800 to 1880) from Lake Bonaparte is 52.5 ng/g. The average mercury concentrations of pre-anthropogenic sediments from Lake Ellen and Cedar Lake (in the UCR within the area influenced by the Trail smelter) are 46.9 and 50.8 ng/g. All three of these values are consistent with the expected range of background values for mercury concentrations in sediments and mineral soils as reviewed below.

Majewski et al (2003) measured Hg concentrations in fine-grained sediments from uncontaminated areas of the Upper Columbia River Valley and found mercury concentrations ranging from 10 to 70 nanograms/gram (ng/g or parts per billion by weight) mercury. Nater and Grigal (1992) studied regional distributions of Hg in upper mineral soils across north-central USA and found range from 15-30 ng/g. In study of Hg concentrations in forest soil profiles in Central Washington State, Biswas et al (2008) found Hg concentrations in mineral soils u to a maximum of 40 ng/g and in a study of Wyoming forest soils found Hg concentrations in mineral soils up to 45 ng/g (Biswas et al 2007). The Provincial Government of British Columbia provides current guidelines for "Regional Background Soil Quality Estimates for Inorganic Substances" for its 7 Provincial regions. Region #4 "Kootenay" includes the Upper Columbia River in BC and the background value determined for that region is 25 ng/g (BCME 2010). An finally, the Washington State Mercury Chemical Action Plan (WSMCAP, 2003) reviews evidence for background soil and sediment Hg concentrations by region and provides a regional background value of 7 to 43 ng/g for regions across the State of Washington. Thus the average background values for pre-anthropogenic sediment from Lakes Bonaparte, Cedar and Ellen fall within the expected range of background mercury concentrations.

The sediment Hg concentration profiles for the three studied lakes are plotted in Figure 1. In order to focus o the anthropogenic atmospheric mercury flux to the lakes we subtracted the average pre-anthropogenic mercury concentration for each lake (52.5 , 46.9 and 50.8 ng/g for Lake Bonaparte, Ellen and Cedar, respectively) from the post-1880 core samples. The anthropogenic mercury concentrations were then converted to annual fluxes by multiplying concentrations by the average sediment accumulation rate (determined from ^{210}Pb dating; Vlassopoulos 2014) for each core sample and this is plotted against their ^{210}Pb age on Figure 2. The Lake Bonaparte background anthropogenic mercury flux rises above pre-anthropogenic background levels in approximately 1900 and rises to a maximum of 2.3 µ m^{-2} yr^{-1} in the mid 1960s. Van Metre (2012) compiled data on mercury fluxes estimated from studies of remote lake sediment cores in the western USA and found that mercury fluxes rose by 3 to 8 µ m^{-2} yr^{-1}

between 1900 and the 1960s. In the 1960s the anthropogenic mercury flux at Lake Bonaparte was similar to both Todd Lake (Oregon) and Lake Elbert (Colorado), but was considerably lower than at Hobbs Lake (Wyoming). This demonstrates that Lake Bonaparte is in low mercury deposition zone; it has very few mercury point sources in the upwind (western) direction (National Atmospheric Deposition Network, NADP 2014) and it serves as an excellent reference lake for this study.

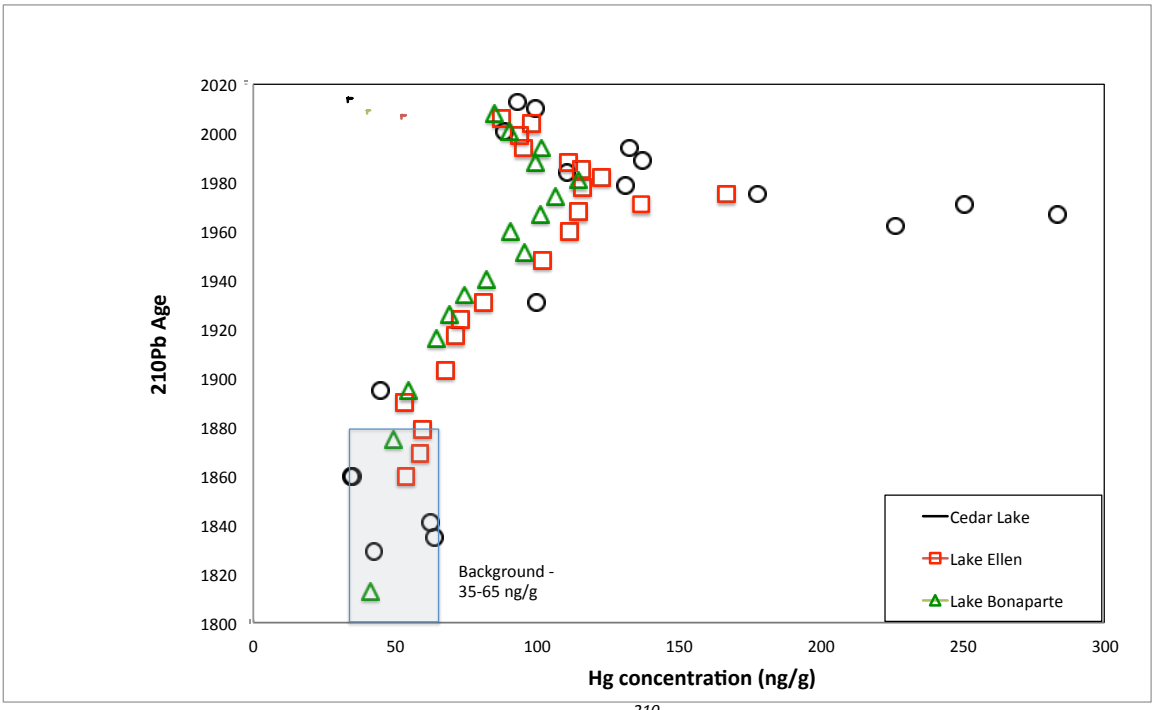

Figure 1: The Hg concentration in ng/g plotted against the ^{210}Pb age of each sediment slice in cores from Lake Bonaparte, Lake Ellen and Cedar Lake in Washington State. Samples from the 1800 to 1880 time interval are used to estimate the pre-anthropogenic background Hg concentration in the lake sediments.

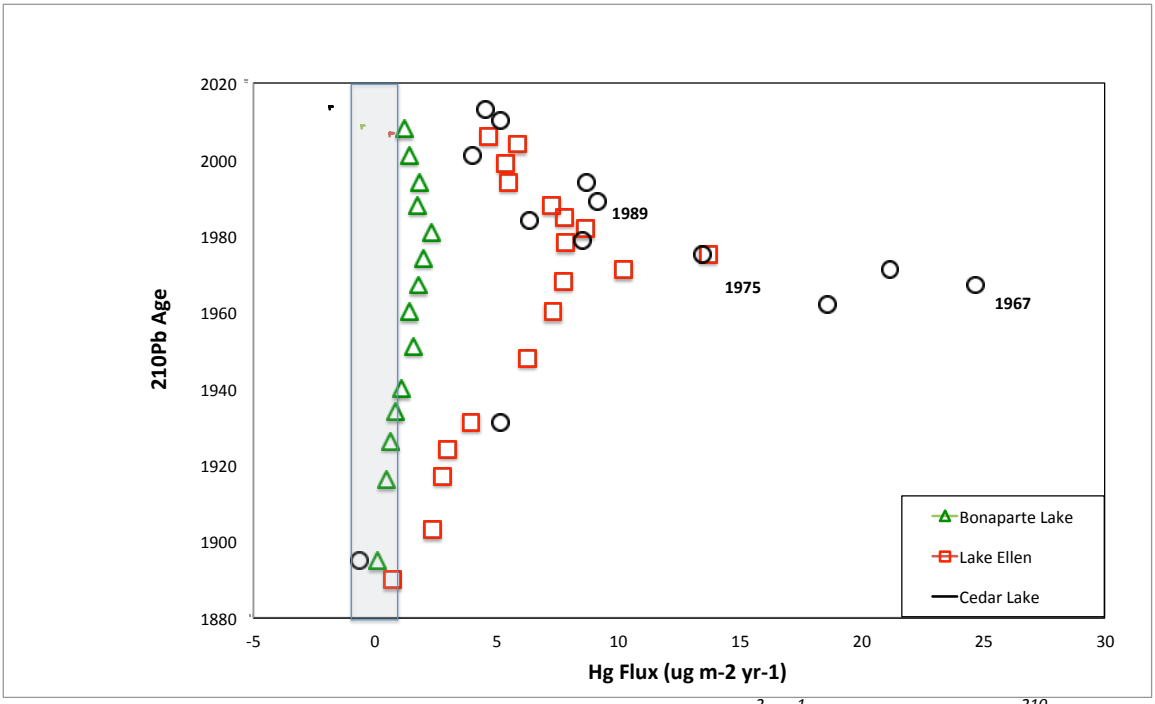

Figure 2: The estimated anthropogenic flux of Hg to the lakes in µg m^{-2} yr^{-1} plotted against the ^{210}Pb age of each sediment slice in cores from Lake Bonaparte, Lake Ellen and Cedar Lake in Washington State. Years are labeled for peak concentrations but they have an uncertainty of about 10 years.

MERCURY FLUXES IN PROXIMAL AND INTERMEDIATE DISTANCE LAKES

Cedar Lake and Lake Ellen were chosen to provide a spatial gradient in metal deposition from the Trail smelter. The peak anthropogenic mercury flux for each lake occurred in the 1960s. As discussed above, Lake Bonaparte records a peak flux of 2.3 µ m^{-2} yr^{-1} in in the mid 1960s. Lake Ellen records peak flux of 13.7 µ m^{-2} yr^{-1} in the mid 1960s, and Cedar Lake records a peak flux of 24.7 µ m^{-2} yr^{-1} also in the mid 1960s. Mercury fluxes decreased in all three lakes following the 1960s and in the 2000s the three lakes recorded fluxes of 1.2, 5.2 and 5.8 µ m^{-2} yr^{-1} in Lakes Bonaparte, Ellen and Cedar, respectively. The magnitude of the peak mercury fluxes in the three lakes correspond to their proximity to the Trail smelter and to the pattern of forest damage from Trail emissions (USDA 1936). Mercury is generally considered to be quite immobile in lake sediments but we cannot rule out minor mobility of mercury in the sediments via pore water and bioturbation, which might cause concentration peaks to be less sharp. Summing over the entire sediment record from 190 to the present yields an estimate of total atmospheric input of anthropogenic Hg of 135, 600 and 980 µ m^{-2} yr^{-1} in Lakes Bonaparte, Ellen and Cedar, respectively. Multiplying by the area of the lakes yields an estimate that 80, 180, and 190 g of anthropogenic Hg was added to Lake Bonaparte, Lake Ellen and Cedar Lake, respectively between 1900 and the present. The topographically and meteorologically controlled distribution of atmospheric pollutants from Trail would not be expected to differ substantially between the 1930s and the periods of Trail Smelter operations before and after this time period (Haney 2011). Vlassopoulos (2014) reports increases in the lead concentration of sediments in Lake Bonaparte, Lake Ellen and Cedar Lake beginning in about 1900 that mimic the patterns of Hg concentration. Measurement of Pb isotopic compositions in these sediments demonstrates that

they are derived from Trail smelter emissions and excludes local ores and other smelters processing local ores (Vlassopoulos 2014).

Several approaches in addition to analyses of lake sediments have been used to estimate Hg deposition fluxes across the United States and these provide a useful comparison with the values we measured for background deposition in Lake Bonaparte in the 2000s. The first approach uses data on annual wet deposition of mercury at National Atmospheric Deposition Program sites across the country and extrapolates values based on precipitation data. This approach yields mercury deposition estimates for 2011 of 1-4 μg m^{-2} yr^{-1} in the vicinity of Lake Bonaparte (NADP 2014). A second approach uses air pollution models based on known emission sources and deposition processes and one such study by researchers at the US-EPA estimated a mercury deposition rate of 0.3 to 3 μg m^{-2} yr^{-1} in the vicinity of Lake Bonaparte (Bullock 2000). These mercury deposition estimates are only rough approximations, but they demonstrate that our lake sediment derived mercury flux estimates are in the same range of values estimated by other methods. Using Lake Bonaparte as a baseline we see that Lake Ellen and Cedar Lake received peak mercury deposition of 6 and 11 times the regional background fluxes in the 1960s.

TIME SERIES OF TRAIL SMELTER MERCURY DEPOSITION FROM LAKE CORES

The time series of mercury flux to Cedar Lake and Lake Ellen gives an estimate of how mercury deposition from the Trail smelter changed through time at the locations of these lakes during the time of smelter operation. The mercury flux to both lakes rose above the background established by Lake Bonaparte in about the year 1900, which is consistent with the smelter first going online in 1895. The sediment Hg concentration records in Cedar Lake and Lake Ellen provide information on the relative magnitude of post 1900s Hg deposition to the lakes, but it is difficult to relate these fluxes quantitatively to annual Trail smelter total mercury emissions. This is because the deposition of mercury from the atmosphere is complex and involves both chemical and physical processes. Mercury is emitted to the atmosphere in three operationally defined chemical forms (Schroeder and Munthe 1998): gaseous elemental mercury (GEM), reactive gaseous mercury (RGM) and particulate bound mercury (PBM). The proportion of mercury emitted in each of these forms is not known for the Trail smelter and would be expected to vary within each particular industrial process occurring at the smelter, the operating conditions of the plant, the chemistry of the materials being processed (in particular the halogen content), and the use of various emissions control devices. Once emitted, RGM is known to deposit rapidly in raindrops and snow crystals, GEM can either enter the global atmospheric pool or be oxidized to RGM in the atmosphere, and PBM deposition varies depending on the particle sizes with which mercury is associated. The rates of GEM oxidation to RGM depend on sunlight and the abundance of chemical oxidants such as ozone and halogens. Once GEM is oxidized to RGM it is rapidly deposited in precipitation. Thus, due to the complexities of mercury transport, chemical reaction and deposition from the atmosphere, it would be difficult to quantitatively correlate fluxes of mercury to Lake Ellen and Cedar Lake with total mercury emissions from the Trail smelter through time.

We have, however, measured fluxes directly using lake sediment analyses and we find that the mercury flux records for Lake Ellen and Cedar Lake can be related qualitatively to Trail emissions and show substantial fluxes through the 1970s. Reductions in mercury fluxes to the lakes are apparent during the 1980s and 1990s during a time period when actions were taken to reduce

mercury emissions through implementation of emissions control processes in various parts of the Trail facility. Finally, mercury fluxes to all three study lakes appear to have stabilized at a reduced value in the 2000s and 2010s. Queneau (2014) summarized Hg emissions estimates from the Trail smelter between the 1980s and the 2000s and finds a drop in emissions that shows the same general trend as the lake sediment core records over this time interval.

3.2 **OPINION 2**

Mercury stable isotope ratios in sediments from Lake Bonaparte, Lake Ellen and Cedar Lake provide isotopic fingerprints of changing proportions of mercury between ore deposits (or smelters that process the ores) and regional/global background atmospheric sources. This demonstrates that mercury in Cedar Lake sediments is derived primarily from ore deposit (or smelter) sources, and that about half of the mercury in Lake Ellen sediments is derived from ore deposit (or smelter) sources.

OVERVIEW OF THE THREE TIME PERIODS IN LAKE MERCURY ISOTOPE RATIO RECORD

Variations in the ratios of the isotopes of mercury in sediments from Lake Bonaparte, Lake Ellen and Cedar Lake can be best understood by dividing the lake sediment record into three time periods. 1) Pre-1900 sediments have low mercury concentrations and the mercury isotope ratios record varying local geological influences on the sediment generated within each lake catchment. 2) Post-1900 mercury concentrations rapidly increased and peaked in the 1950s and 1960s. The mercury isotope ratios during these peak periods represent mixing between global/regional sources of mercury (mainly from coal combustion) and a local point source of mercury consistent with an ore deposit or smelter source. 3) In the 1970s through the 1990s mercury concentrations in the lake sediments diminished and by the 2000s the mercury isotope ratios reflect mixing between a global/regional source of mercury (mainly from coal combustion) and a diminished, but still significant, point source of mercury from an ore deposit or smelter source

MERCURY ISOTOPE NOMENCLATURE

The element mercury (which has 80 protons) exists with seven different numbers of neutrons (isotopes) and differs ranges in mass between 196 and 204 atomic mass units. The ratios between these isotopes of mercury vary in nature in two fundamentally different ways: due to mass dependent fractionation (MDF) and due to mass independent fractionation (MIF) (Bergquist and Blum 2007). Following common practice we reference mercury isotopic compositions to the National Institute of Standards and Technology Standard Reference Material (NIST-SRM) 3133 and use two measures of the isotopic composition: δ^{202}Hg ("delta 202") for MDF and Δ^{199}Hg ("cap delta 199") for MIF in permil (‰) (see Blum and Bergquist 2007). δ^{202}Hg is the difference in ^{202}Hg/^{198}Hg between the NIST-3133 standard and a sample in units of permil (‰), and Δ^{199}Hg is the difference in ^{199}Hg/^{198}Hg between a sample and the ^{199}Hg/^{198}Hg ratio predicted by mass dependent isotope fractionation, also in units of ‰. In samples of modern day atmospheric deposition and in ore deposits analyzed from around the globe there is a wide range of values of δ^{202}Hg due to chemical reactions that produce mass dependent fractionation (Blum et al 2014). However, in both atmospheric and ore deposit samples analyzed

from around the globe there is a narrow and distinct range of Δ^{199}Hg values because MIF is produced only by the magnetic isotope effect (photochemical reactions involving short-lived radical pairs) and the nuclear volume effect (which is unusual in natural systems and of very small magnitude) (Bergquist and Blum 2009). In a series of previous studies the lack of MIF (Δ^{199}Hg=0) in mercury from ore deposits globally, and mercury produced from these ore deposits and used in industry, has been used to identify the addition of mercury to natural environments from anthropogenic sources (Estrade et al 2011; Sonke et al 2010; Kwon et al 2014; Mil-Homens et al 2013; Foucher et al 2013; Ma et al 2013).

In the analyses of mercury isotopes from lake sediments reported here we observe a large variation of δ^{202}Hg in pre-anthropogenic sediments (Appendix B) in a time period when δ^{202}Hg was controlled by the local geology of each lake catchment. The isotopic variability then narrows for the lake sediments as they become influenced by anthropogenic sources, leading to δ^{202}Hg values that overlap between the three lakes and leading to a situation where the δ^{202}Hg value is not diagnostic of the mercury source to the lakes. In contrast, Δ^{199}Hg values (MIF) are non-overlapping between the three lakes during the time of anthropogenic influence and are diagnostic of the varying importance of different mercury sources to the three lakes. Because of the better diagnostic power of the Δ^{199}Hg value in this situation we focus on the use of this parameter in our discussion of changing mercury sources given below.

TIME PERIOD 1: NATURAL BACKGROUND (pre-1900)

Mercury concentration profiles in Lake Bonaparte, Lake Ellen and Cedar Lake clearly define a transition from pre-industrial times with background Hg concentrations of 50±10 ng/g (1sd) to higher values beginning in approximately the year 1900. This range of background sediment concentrations is consistent with previous estimates of background mineral soil and sediment mercury concentrations in the region, determined by several different methods, as discussed in Opinion #1. Mercury in the pre-1900 lake sediments represents predominantly Hg in geological materials in each lake's watershed. The isotopic composition of this pre-1900 mercury in the three lakes varies within the range previously observed for pre-industrial mercury in rocks, soils and sediments (Blum et al 2014) and is distinct for each of the three lakes (Appendix B), each of which has a different geological bedrock composition (WDNR 2013). It is important to consider that as anthropogenic mercury begins to enter the lakes (in about 1900) the small flux of pre-anthropogenic mercury is overwhelmed by anthropogenic inputs and the isotopic composition becomes dominated by the various anthropogenic sources.

TIME PERIOD 2: PEAK LOCAL INPUTS (1950s AND 1960s)

Mercury concentration profiles in Lake Bonaparte, Lake Ellen and Cedar Lake all increase above background around 1900 and continue to increase steadily with maxima in the 1960s (Figure 1). Δ^{199}Hg values of sediment core samples from the 1950s and 1960s are plotted versus 1/Hg concentration as filled symbols on Figure 3. The reciprocal of Hg concentration is plotted following common practice on Figure 3 to linearize mixing relationships on this diagram. The Cedar Lake samples plot within 2σ analytical uncertainty of Δ^{199}Hg=0 (-0.04 to 0.06‰), which is the expected Δ^{199}Hg value of mercury derived from ore deposits across the globe and is thus the likely composition emitted by the Trail smelter. Thus we conclude that Hg deposited to Cedar

Lake in the 1950s and 1960s was predominantly derived from ore deposits rather than the regional/global background. This is consistent with origin of Hg from the nearby Trail smelter. As mentioned above, Pb isotope data in the lake sediments preclude local ores or smelters as the source of metals in these lakes (Vlassopoulos 2014). The background control lake, Lake Bonaparte, has elevated Δ^{199}Hg ranging from 0.32 to 0.42‰, which is in the range expected for regional/global derived mercury currently deposited in precipitation in the mid-continent of North America.

Δ^{199}Hg values of individual precipitation events that have been measured are all positive, but vary in magnitude between rainfall events due to changes in sources and atmospheric conditions. To average out variability from individual precipitation events it is instructive to use mean values of rainfall events at each site, and this provides the best estimate of the average precipitation Δ^{199}Hg values, and thus the annual average Δ^{199}Hg that would enter a lake in precipitation. Three studies that have been done of Δ^{199}Hg in precipitation in rural sites in northern North America found mean Δ^{199}Hg values of 0.30 ±0.13‰ in the Great Lakes region (Gratz et al; 2010), 0.59 ±0.25‰ in northern Ontario (Chen et al 2012) and 0.48 ±0.27‰ in northern Wisconsin (Demers et al 2013). The average of these values (0.46‰) provides the best estimate of the regional/global atmospheric Δ^{199}Hg value in North America. Thus we conclude that Hg deposited to Lake Bonaparte in the 1950s and 1960s was predominantly derived from regional/global sources in precipitation (Figure 3). A simple mixing calculation indicates that the isotopic composition of Lake Ellen sediments is derived approximately 1/2 from mercury originating from the Trail smelter and approximately 1/2 from mercury of regional/global atmospheric sources. Mercury in Cedar Lake in the 1960s has an isotopic composition indistinguishable from ore deposit (or smelter) values.

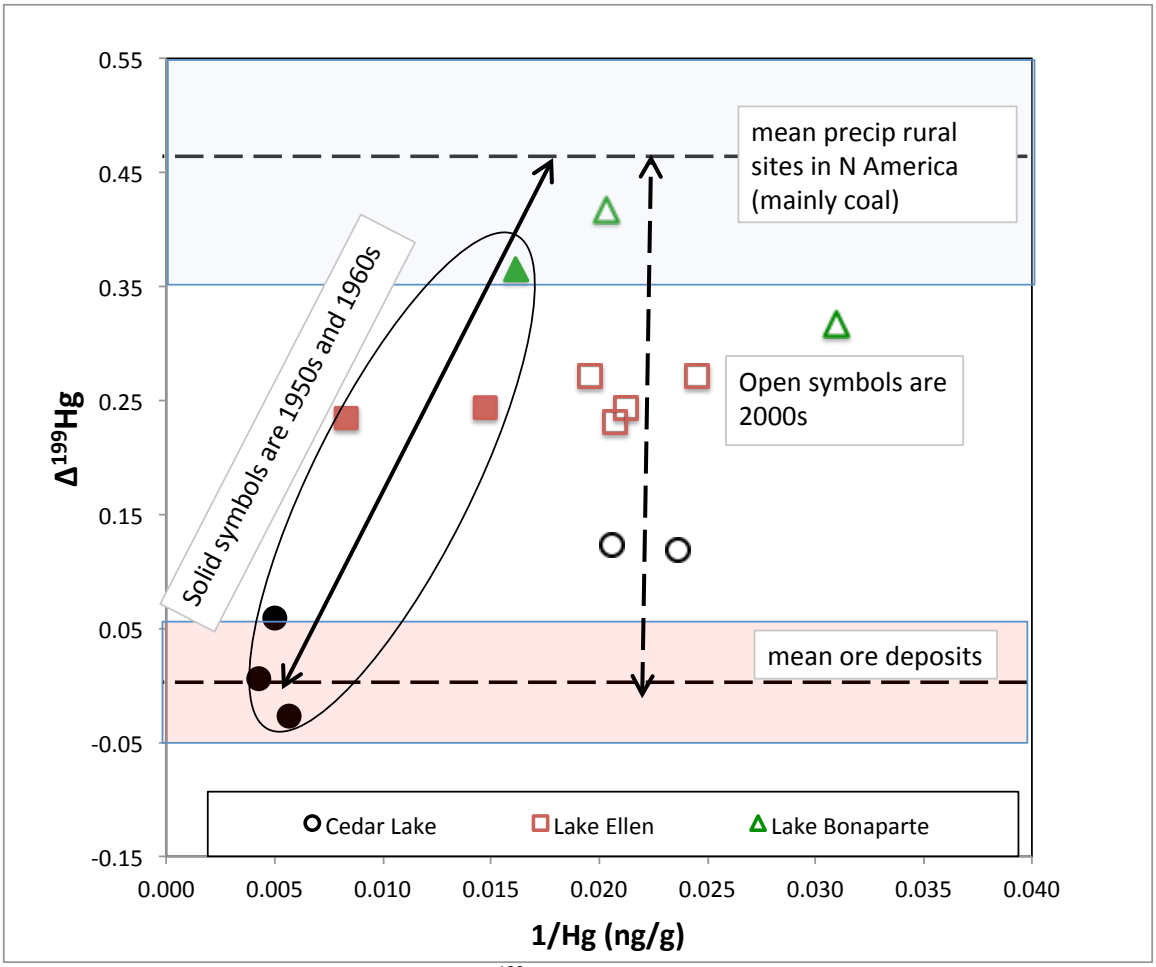

Figure 3: The mercury isotopic composition (Δ^{199}Hg) in permil (‰) versus the reciprocal of the mercury concentration in ng/g. Note that mixtures of endmembers (depicted as arrows) plot as lines on this diagram and that mercury concentrations decrease to the right on the X-axis. Analytical uncertainty for measurements of Δ^{199}Hg is ±0.06‰ (2σ). The blue and red shaded areas indicate the 1σ range of North American precipitation and global ore deposits, respectively.

TIME PERIOD 3: DIMINISHED LOCAL INPUT (2000s)

Mercury concentration profiles in Lake Bonaparte, Lake Ellen and Cedar Lake all decrease from peaks in the 1960s and converge on a common value of about 90 ng/g Hg in the 2000s (Figure 1). The Δ^{199}Hg values of Cedar Lake shift significantly from 0.0‰ to a value of 0.12 ‰, which is a shift in the direction of the background atmospheric value of Lake Bonaparte and indicates a significant decrease in the proportion of total Hg input coming from the Trail smelter. In contrast, the Δ^{199}Hg value of Lake Bonaparte (which is representative of the regional background Hg) remains constant over this time interval. The value of Lake Ellen shifts to a slightly higher Δ^{199}Hg value indicating that there is a decrease in the proportion of total Hg input coming from the Trail smelter. Mercury isotopic analyses of core samples from the 2000s are plotted as open symbols on the plot of Δ^{199}Hg versus 1/Hg concentration (Figure 3) to illustrate the changes in both Δ^{199}Hg value and Hg concentration between the 1950-1960s and the 2000s. In the 1950-1960s data from the three lakes fall along the solid arrow on Figure 3 whereas during the 2000s

data from the three lakes fall along the dashed arrow on Figure 3. This data indicates that in the 1950-1960s Cedar Lake received essential all of its mercury input from the Trail smelter, and that this had shifted by the 2000s to Cedar Lake receiving approximately 2/3 of its mercury from Trail and 1/3 from regional/global sources.

IMPLICATIONS OF OPINION 2

The mass independent fractionation of mercury isotopes (Δ^{199}Hg) provides a unique means for fingerprinting and separating the inputs of mercury to lakes from global/regional background deposition (mostly from global coal combustion) compared to input to lakes from local point sources of mercury from Trail smelter stack emissions. The results are consistent with those ascertained from investigation of mercury concentration profiles in lakes of variable distances from the Trail smelter. The isotope results validate the conclusions reached from analysis of Hg concentration patterns and lead isotope ratios (Vlassopoulos 2014) in the lake sediments by providing an independent means of differentiating between Trail smelter sources and regional/global sources of mercury deposition.

3.3 **OPINION 3**

Historical data concerning Trail smelter SO_2 emissions and meteorological analysis of the transport of its emissions corroborates and provides independent evidence for my opinions identifying atmospherically deposited metals from the Trail smelter in the Upper Columbia River Valley site.

There is a clear historical record of SO_2 emissions from the Teck Trail smelter and the transport of this SO_2 into the Upper Columbia River Valley (Queneau 2011; Haney 2011). Evidence for damage to vegetation by the Trail SO_2 plume was documented during the years 1929-1931 when maps of the zone of visible smelter plume injury to forest vegetation were made by the USDA and these maps show damage extending ~80 km south from Trail to the town of Kettle Falls (USDA 1936). In 1941 the International Joint Commission (IJC) also substantiated direct SO_2 damage to agricultural crops and commercial forests in the Northport area from the Trail smelter SO_2 plume (IJC, 1941).

Heavy metals and SO_2 have been emitted simultaneously from the Teck Trail smelter throughout its time of operation (Queneau 2011). Whereas SO_2 is known to produce visible evidence of the geographic distribution of its deposition, heavy metal deposition does not produce visible signs of deposition. However, it is well known that when SO_2 and heavy metals are emitted together from stacks, they are transported and deposited by the same meteorological processes, and therefore they are co-deposited. Landis (2014) reviewed the current literature on SO_2 and heavy metal co-deposition and summarized recent studies that clearly demonstrate SO_2 and heavy metal co-deposition from high-temperature metal processing plants such as the Teck Trail smelter. Landis (2014) also summarized studies that show in some cases that SO_2 and heavy metals are transported and deposited on the same aerosol particles.

The predominant wind direction in the UCR follows the Columbia River Valley and often drains to the south from Trail during the night and early morning (Haney 2011) providing a means for transporting smelter emissions to the south. Landis (2014) summarized studies that conclude

there is about a 40 km range of substantial heavy metal soil and lake contamination around other smelter operations that have been studied. Landis (2014) also summarized studies that indicate that SO_2 can effectively be used as a tracer of heavy metals emissions from smelting activities and, therefore, it follows that heavy metal deposition likely impacted the UCR concurrently with the documented SO_2 damage events that extend up to 80 km to the south from the Teck Trail facility.

There is limited air quality monitoring data from 1992 to 1998 at sites at (b)(6) and Northport Elementary School in the UCR (WSDOE, 1999). Landis (2014) considered chemical data for total suspended particulates from this study and found that zinc and lead concentrations were correlated at the two sites, that the concentrations were higher at the site closer to Trail, and that the zinc/lead ratios during periods of high metal concentrations matched ratios from furnace stack emissions at the Teck Trail smelter. Together these data are consistent with transport and deposition of metals from the Teck Trail facility into the UCR Valley.

Evidence for transport of SO_2 from Trail into the UCR Valley, evidence for co-deposition of SO_2 and heavy metals at other sites where detailed investigations have been completed, and chemical analyses of total suspended atmospheric particles in the UCR Valley corroborate and provide independent evidence for Opinions 1 and 2 which identify the deposition of heavy metals from the Trail smelter in the Upper Columbia River Valley.

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APPENDIX A

JOEL D BLUM
PUBLICATIONS OF PAST 10 YEARS student advisee

2014 (n=185; ISI h=43; Google h=52, citations=8338)

Li, M., Sherman, L.S., Blum, J.D., Grandjean, P., Mikkelsen, B., Weihe, P., Sunderland, E.M. and Shine, J.P. Assessing sources of human methylmercury exposure using stable mercury isotopes. *Env. Sci and Tech.* (in press)

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APPENDIX B: Data Tables

Cedar Lake, Washington								
Sample #	Hg ng/g	Hg ng/g	1/Hg	δ^{202}Hg	Δ^{199}Hg	Δ^{201}Hg	Mean	^{210}Pb
CDR3-		minus bkgd	minus bkgd				depth (cm)	Age
0001	93.1	37.0	0.027	-0.86	0.12	0.23	0.5	2013
0203	99.4	43.3	0.023	-0.79	0.12	0.09	2.5	2010
0405	88.3	32.2	0.031				4.5	2001
0607	132.7	76.6	0.013				6.5	1994
0708	137.0	80.9	0.012				7.5	1989
0809	110.6	54.5	0.018				8.5	1984
0910	131.2	75.1	0.013				9.5	1979
1011	177.7	121.6	0.008				10.5	1975
1112	250.3	194.2	0.005	-0.92	0.06	0.04	11.5	1971
1213	283.5	227.4	0.004	-0.96	0.01	0.06	12.5	1967
1314	226.0	169.9	0.006	-0.82	-0.03	0.01	13.5	1962
1617	99.5	43.4	0.023				16.5	1931
1819	44.5	-11.6	-0.086				18.5	1895
2022	34.4	-21.7	-0.046				21.0	1860
2426	62.1	6.0	0.168	-0.51	-0.06	-0.11	25.0	1841
2628	63.8	7.7	0.130	-0.54	-0.09	-0.15	27.0	1835
2830	42.5	-13.6	-0.073				29.0	1829
3234	45.4	-10.7	-0.093				33.0	
3638	54.6	-1.5	-0.652				37.0	
4042	28.7	-27.4	-0.036				41.0	
4446	42.6	-13.5	-0.074				45.0	
4850	43.4	-12.7	-0.079	-0.84	-0.11	-0.10	49.0	
5254	38.5	-17.6	-0.057	-0.83	-0.15	-0.17	53.0	
Mean 1800-1880								
	56.1							
Replicates								
2022	34.6							
4042	26.0							

Lake Ellen, Washington								
Sample #	Hg ng/g	Hg ng/g	1/Hg	δ^{202}Hg	Δ^{199}Hg	Δ^{201}Hg	Mean	^{210}Pb
ELN-2		minus bkgd	minus bkgd				depth (cm)	Age
0004	87.7	40.8	0.025	-0.74	0.27	0.16	2.0	2006
0405	98.0	51.1	0.020	-0.85	0.27	0.17	4.5	2004
0607	94.0	47.1	0.021	-0.71	0.24	0.19	6.5	1999
0809	95.0	48.1	0.021	-0.83	0.23	0.17	8.5	1994
1011	110.7	63.8	0.016				10.5	1988
1112	115.3	68.4	0.015				11.5	1985
1213	122.4	75.5	0.013				12.5	1982
1314	115.8	68.9	0.015				13.5	1978
1415	166.8	119.9	0.008	-0.90	0.23	0.18	14.5	1975
1516	136.6	89.7	0.011				15.5	1971
1617	114.7	67.8	0.015	-0.87	0.24	0.15	16.5	1968
1819	111.2	64.3	0.016	-0.72	0.31	0.19	18.5	1960
2022	101.9	55	0.018				21.0	1948
2426	81.3	34.4	0.029				25.0	1931
2628	72.9	26	0.038				27.0	1924
2830	71.1	24.2	0.041				29.0	1917
3234	67.8	20.9	0.048				33.0	1903
3638	53.3	6.4	0.156				37.0	1890
4042	59.6	12.7	0.079				41.0	1879
4446	58.7	11.8	0.085				45.0	1869
4850	53.8	6.9	0.145				49.0	1860
5254	42.0	-4.9	-0.204	-1.04	-0.06	0.00	53.0	
5661	33.1	-13.8	-0.072	-1.05	-0.09	-0.18	58.5	
Mean 1800-1880								
	46.9							
Replicates								
2426	80.7							

Lake Bonaparte, Washington								
Sample #	Hg ng/g	Hg ng/g	1/Hg	δ^{202}Hg	Δ^{199}Hg	Δ^{201}Hg	Mean	^{210}Pb
BON-2		minus bkgd	minus bkgd				depth (cm)	Age
0102	84.8	32.3	0.031	-0.83	0.32	0.27	1.5	2008
0203	90.2	37.7	0.027				2.5	2001
0304	101.6	49.1	0.020	-0.64	0.42	0.27	3.5	1994
0405	99.2	46.7	0.021				4.5	1988
0506	114.4	61.9	0.016	-0.71	0.37	0.24	5.5	1981
0607	106.5	54.0	0.019				6.5	1974
0708	100.8	48.3	0.021				7.5	1967
0809	90.8	38.3	0.026				8.5	1960
0910	95.5	43.0	0.023				9.5	1951
1011	82.0	29.5	0.034				10.5	1940
1112	74.4	21.9	0.046				11.5	1934
1213	69.0	16.5	0.061				12.5	1926
1314	64.7	12.2	0.082				13.5	1916
1516	54.6	2.1	0.476	-0.81	0.25	0.12	15.5	1985
1718	49.4	-3.1	-0.323	-1.00	0.25	0.21	17.5	1875
2224	41.2	-11.3	-0.088	-1.14	0.13	-0.03	23.0	1813
2830	25.8	-26.7	-0.037				29.0	
3234	27.0	-25.5	-0.039				33.0	
3638	32.8	-19.7	-0.051				37.0	
4042	28.9	-23.6	-0.042				41.0	
4446	26.5	-26.0	-0.038				45.0	
5254	20.4	-32.1	-0.031				53.0	
Mean 1800-1880								
	52.5							
Replicates								
708	98.2							
2830	24.3							
4446	23.5							